Health Effects of Acid Aerosols on North American Children: Air Pollution Exposures

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Air pollution measurements were conducted over a 1-year period in 24 North American communities participating in a respiratory health study. Ozone, particle strong acidity, sulfate, and mass (PM_{10} and $PM_{2.1}$) were measured in all communities. In 20 of the communities, sulfur dioxide, ammonia, nitric acid, nitrous acid, and particulate nitrate were measured. The sampler was located centrally in the community whenever possible and samples were collected every other day. Concentrations of particle strong acidity, mass, sulfate, and ozone were highly correlated both in the region of the country defined as a high-sulfur source area and in the downwind transport regions. These regions of the eastern United States and southern Canada experienced the greatest particle strong acidity, sulfate, and particle mass concentrations during the spring and summer months (May–September). The particle strong acidity concentrations were highest in regions close to the high sulfur emission areas of the United States; that is, in the area immediately to the west of the Appalachian Plateau and west of the Allegheny Mountains (western Pennsylvania, eastern Ohio, and West Virginia) up through southern Ontario. The frequency of particle strong acidity events decreased with transport distance from the region of highest sulfur emissions. Low particle strong acidity and sulfates were found at the western and midwestern sites of both the United States and Canada. Substantial concentrations of nitric acid were found in two of the California sites as well as many sites in the northeastern portion of the United States. Sites selected for the epidemiologic study provide a range of annual mean particle strong acidity exposures from below the limit of detection to more than 50 nmol/m^3. *Key words*: acid aerosols, air pollution, nitric acid, ozone, particulate matter. *Environ Health Perspect* 104:492–499 (1996)

Although recent epidemiologic studies have suggested that increased morbidity (*1*) and mortality (*2,3*) are associated with exposures to ambient particulates, the chemical or physical property of particulate matter which is responsible for these associations is not clear. Many of the potentially toxic components of the particles, including particle strong acidity (H^+) and many trace metals (e.g., vanadium, manganese, lead, and arsenic), are associated with fine particulates (*4*). Changes in size and chemical composition of particulates could lead to differences in particle toxicity that can vary between sites and over time, for equivalent mass concentrations. In the northeastern United States and southeastern Canada, the highest particle exposures occur during the summer (*5,6*) with sulfate representing a large fraction (40%) of the particle mass. Atmospheric conditions favoring acid aerosol formation also enhance ozone formation. Nitrogen oxide emissions are converted to gaseous acids (such as nitric and nitrous acid) and particle nitrates under conditions conducive to the formation of ozone. Thus the epidemiologic findings of adverse health effects associated with PM_{10} may represent an underlying casual association with some specific characteristic of the particles or effects associated with some other pollutant that is correlated with the mass concentration.

Ambient concentrations of PM_{10}, $PM_{2.1}$, particle strong acidity, sulfate, nitrate, and ammonium, nitric and nitrous acid, sulfur dioxide, and ozone were measured for approximately 1 year in each of 24 communities in the United States and Canada as part of a study of health effects of particle strong acidity (*7*). Respiratory illness and pulmonary function of approximately 15,000 children were measured in these 24 cities. This paper presents the results of the air pollution monitoring in these communities. The results of the respiratory health effects assessments, along with the observed associations with air pollution exposures, are presented by Raizenne et al. (*8*) and Dockery et al. (*9*) in this issue.

Methods

We selected study communities to provide a range of exposures to acid aerosol and ozone, to limit confounding from other air pollutants, and to limit possible confounding by population characteristics. Specifically, communities were targeted with the following characteristics based on 1980 census data: populations between 20,000 and 30,000 for adequate numbers of schoolchildren, racial homogeneity, low poverty levels, high residential stability, and balanced use of gas for cooking. From the demographically acceptable communities, specific sites were selected based on records of ozone, sulfate, acid deposition, and wherever possible particle strong acidity concentrations.

Communities were selected to contrast ambient exposures to ozone and particle strong acidity. We identified three of the four combinations of high and low expected ozone and particle strong acidity conditions. Communities with high particle strong acidity and low ozone concentrations could not be identified. The original study design also called for a third selection dimension based on high and low gaseous acids (nitric and nitrous acid). It was not possible to identify communities with gaseous acid exposures independent of ozone, and this constraint in the design was relaxed.

Eight communities were monitored in the first year of the study (1988–1989), nine in the second year (1989–1990), and seven in the third year (1990–1991). Six communities were in Canada, and 18 were in the United States (Fig. 1). The communities were grouped into four broad geographic clusters to clarify the presentation: high particle strong acidity regions in the east near sources of sulfur emissions (Sulfate Belt) and downwind (Transport Region); high ozone but low particle strong acidity regions in the west (West Coast); and low

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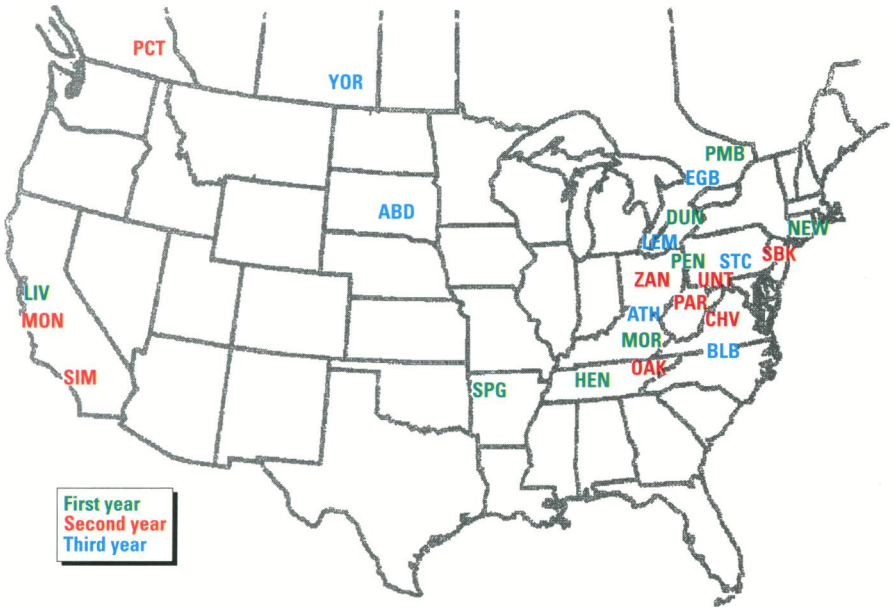

Figure 1. Locations of the study sites by year of study: 24 cities, United States and Canada, 1988–1991. Site codes are provided in Table 1.

Table 1. Distribution of particulate measurements by pollutant and community: 24 communities, United States and Canada, 1988–1991[a]

Region	Site code	No. of samples[b]	Particle strong acidity (nmol/m³) Mean	Max	Sulfate particles (nmol/m³) Mean	Max	Respirable particulate matter (µg/m³) Mean	Max	Inhalable particulate matter (µg/m³) Mean	Max
Sulfate Belt										
Hendersonville, TN	HEN	170	39.3	222	63.5	235	16.4	52	29.5	75
Oak Ridge, TN	OAK	167	43.3	292	65.5	306	17.0	64	23.4	62
Morehead, KY	MOR	172	43.7	302	67.5	285	20.0	76	28.4	75
Blacksburg, VA	BLB	175	33.5	223	65.8	248	16.5	42	23.8	58
Charlottesville, VA	CHV	168	36.8	316	58.5	275	15.1	52	21.2	59
Zanesville, OH	ZAN	164	35.2	364	66.6	390	16.9	54	26.9	83
Athens, OH	ATH	180	43.5	502	72.8	449	17.3	80	26.0	98
Parsons, WV	PAR	180	48.4	391	63.0	352	17.8	67	25.8	92
Uniontown, PA	UNT	179	51.9	797	75.0	536	20.7	109	29.6	115
Penn Hill, PA	PEN	162	35.2	396	77.3	444	19.3	71	32.7	117
State College, PA	STC	171	40.6	337	64.8	307	13.6	57	19.1	65
Transport Region										
Leamington, ON	LEM	179	19.1	276	48.3	345	14.3	55	23.5	77
Newtown, CT	NEW	165	25.7	186	47.1	271	13.8	56	21.2	77
Egbert, ON	EGB	173	6.3	123	40.0	320	9.7	52	17.1	69
Pembroke, ON	PMB	152	20.6	272	33.9	290	10.4	53	15.4	62
Dunnville, ON	DUN	158	28.9	284	61.5	318	16.2	64	20.2	80
South Brunswick, NJ[c]	SBK	49	48	222	71.7	254				
West Coast										
Simi Valley, CA	SIM	175	15.9	115	32.4	154	18.1	62	31.6	72
Livermore, CA	LIV	175	11.9	39	15.0	79	15.3	90	26.1	82
Monterey, CA	MTY	178	9.9	32	13.3	55	9.0	53	23.6	59
Background										
Springdale, AR	SPG	160	12.7	53	36.3	133	14.4	44	28.5	87
Aberdeen, SD	ABD	175	1.7	19	20.1	213	6.9	30	18.9	72
Yorkton, SK	YOR	163	0.0[d]	10	9.0	45	5.8	37	16.5	47
Penticton, BC	PCT	174	8.5	28	6.8	33	9.3	36	17.3	46

[a]Minimum value in all communities was less than the limit of detection.
[b]Maximum sample size for any of the four parameters.
[c]South Brunswick, NJ sampling was for 21 June 1989–25 August 1989.
[d]Mean particle strong acidity level below limit of detection; set to zero.

pollution regions in central United States and Canada (Background).

Ambient air pollution concentrations were measured for approximately 12 months in each community. Ambient ozone concentrations were measured continuously in each community or at a location representative of ozone concentrations in the general area. Samples of other pollutants were collected every other day using the Harvard/EPA Annular Denuder System (HEADS) sampler (*10–12*). Thompson and co-workers (*13*) have shown that this sampling frequency is sufficient to characterize the annual distribution of particle strong acidity, sulfate, and particle mass concentrations.

In South Brunswick, New Jersey, HEADS samplers were operated by investigators from the Environmental and Occupational Health Sciences Institute, University of Medicine and Dentistry of New Jersey-Robert Wood Johnson Medical School during the summer before administering health questionnaires but not in other months. In Egbert, Ontario, the sampling site was operated by Atmospheric Environment, Canada. Delays in establishing a laboratory in Egbert resulted in only a partial year of sampling for particle mass and particle strong acidity, but a full year of ozone measurements.

The HEADS sampler consists of a glass impactor inlet, two annular denuders, and a filter pack. The inlet removes particles larger than 2.1 µm aerodynamic diameter. The first denuder, coated with sodium carbonate, collects sulfur dioxide (SO_2), nitrous acid (HNO_2), and nitric acid (HNO_3) gases. The second denuder, coated with citric acid, collects ammonia (NH_3). Fine particles ($PM_{2.1}$) are collected on a Teflon filter. Backup sodium carbonate and citric acid-coated glass filters collect acidic gases and ammonia volatilized from the collected fine particles.

Denuders and filters were disassembled and extracted in an ammonia-free hood. Extracts were analyzed by ion chromatography for nitrate, nitrite, sulfate, and ammonium to determine concentrations of nitrous acid (HNO_2), nitric acid (HNO_3), sulfur dioxide (SO_2), and ammonia (NH_3) gases, and particle nitrate (NO_3^-), sulfate (SO_4^{2-}), and ammonium (NH_4^+) ions. H^+ was determined by pH analysis of the Teflon filter extract (*14*).

Fine particle mass concentration ($PM_{2.1}$) was determined using a separate sampler consisting of the same glass impactor inlet used in the HEADS sampler with particles collected on a Teflon filter (*11*). Inhalable particles (PM_{10}) were collected using a similar sampler with two

identical impactor stages which provide a sharp cut at 10 μm aerodynamic diameter (*15*). Total mass of particles collected in both cases were determined gravimetrically.

In two sites with expected low particle strong acidity concentrations (Aberdeen, South Dakota, and Yorkton, Saskatchewan), samples were collected using the Harvard Impactor sampler with an aluminum honeycomb ammonia denuder (*14*) to measure particle strong acidity, sulfate, and ammonium. Collocated sampling has shown good agreement in particle strong acidity measurements of this sampler compared to the HEADS sampler (*16*).

Results

The particle and ozone measurements are summarized in Tables 1 and 2, respectively, and the rest of the measured gaseous pollutants are shown in Table 3. As previously mentioned, the communities were grouped into four categories. The Sulfate Belt signifies those sites that are in the Appalachian region including Ohio, Pennsylvania, Virginia, West Virginia, Tennessee, and Kentucky. Many of the country's largest sulfur emission sources are located in these states (*17*), and previous studies have shown that the region to the west of the Appalachian Plateau and the Allegheny Mountains experiences the greatest amount of acid deposition and the highest sulfate particle concentrations (*18*).

The Transport Region includes communities that experience the effects of sulfate particle pollution transported from source regions, but mean annual concentrations of sulfate and acid particles are uniformly lower than in the Sulfate Belt. These communities are north, northeast, and east of the states with substantial sulfur emissions and experience less frequent episodes of high particle strong acidity events. The West Coast group consists of three California communities with a range of air pollution: Livermore and Simi Valley have higher ozone and nitric acid concentrations, while Monterey was chosen to be a "clean" coastal site. The Background group consists of four continental communities: Springdale, Arkansas; Aberdeen, South Dakota; Yorkton, Saskatchewan; and Penticton, British Columbia.

Particulate Matter

A summary of the particulate measurements PM_{10}, $PM_{2.1}$, sulfate, and particle strong acidity concentrations is shown in Table 1. Annual PM_{10} concentrations for the 24 communities studied ranged from 15 to 32 μg/m^3. The highest PM_{10} concentrations were found at Penn Hills, Pennsylvania, and Simi Valley, California.

Table 2. Distribution of ozone concentrations by parameter and community: 24 communities, United States and Canada, 1988–1991

		Ozone (ppb)					
		Maximum hourly		Daytime[a]		24-hr average	
Region	Samples	Mean	Max	Mean	Max	Mean	Max
Sulfate Belt							
Hendersonville, TN	365	54.4	124	46.9	111	34.3	84
Oak Ridge, TN	365	45.1	86	34.7	66	27.9	58
Morehead, KY	365	53.7	130	45.1	112	34.6	87
Blacksburg, VA	362	51.9	98	44.6	90	31.9	74
Charlottesville, VA	350	49.8	127	43.0	110	29.4	65
Zanesville, OH	365	47.2	141	36.9	97	29.2	66
Athens, OH	364	45.7	122	37.9	104	23.4	76
Parsons, WV	365	49.7	107	42.1	85	30.1	68
Uniontown, PA	365	44.9	114	35.9	96	25.5	72
Penn Hill, PA	364	38.9	153	30.3	116	19.4	71
State College, PA	367	48.4	120	41.5	102	32.8	80
Transport Region							
Leamington, ON	367	45.3	136	37.3	104	29.8	83
Newtown, CT	366	49.4	214	38.5	151	24.8	99
Egbert, ON	365	42.0	103	36.0	84	29.7	72
Pembroke, ON	335	35.8	103	29.5	86	24.7	86
Dunnville, ON	365	50.2	149	41.0	133	32.9	99
South Brunswick, NJ	365	47.3	151	36.9	116	22.6	84
West Coast							
Simi Valley, CA	365	72.5	170	60.4	144	34.8	77
Livermore, CA	365	49.7	150	37.2	89	21.3	44
Monterey, CA	364	40.7	110	34.6	87	28.5	57
Background							
Springdale, AR	348	46.8	94	40.4	86	29.5	70
Aberdeen, SD[b]	125	46.5	71	42.1	67	33.0	57
Yorkton, SK	365	26.9	58	21.0	47	16.3	40
Penticton, BC	365	33.5	62	28.7	53	21.5	43

[a]1000–1800 hr.
[b]Aberdeen sampling for 29 May–12 September 1991.

The highest-single day PM_{10} concentration was 117 μg/m^3, recorded at Penn Hills. In all but three communities (Livermore and Monterey, California, and Penticton, British Columbia), the mean summer (May–September) concentrations of PM_{10} were higher than the annual mean.

Annual mean $PM_{2.1}$ concentrations ranged from 6 μg/m^3 in Yorkton, Saskatchewan, to 21 μg/m^3 in Uniontown, Pennsylvania (Table 1). The eastern communities experienced higher concentrations over the summer months. The three California sites, along with the sites in the western Canadian provinces did not show distinct seasonal patterns. Table 1 includes the maximum 24-hr $PM_{2.1}$ concentrations (μg/m^3) in all 24 sites.

Over the 24 communities, the annual and summer means of PM_{10} and $PM_{2.1}$ were significantly correlated (0.77 and 0.80). The portion of mean PM_{10} in the fine fraction ($PM_{2.1}/PM_{10}$) varied from 35% in Yorkton, Saskatchewan to 80% in Dunnville, Ontario. There were eight communities where the ratio of mean $PM_{2.1}/PM_{10}$ was approximately 0.70 or higher; these communities lined up along the axis of the Appalachian Mountains stretching from Oak Ridge, Tennessee, through western Pennsylvania and southern Ontario. The six communities bordering this area had the next highest $PM_{2.1}/PM_{10}$ ratios. Livermore and Simi Valley, California, had ratios of 0.59 and 0.57, respectively. Other communities had lower $PM_{2.1}/PM_{10}$ ratios, with the "cleaner" communities of Monterey, California; Aberdeen, South Dakota; and Yorkton, Saskatchewan, among the lowest, 0.38, 0.37, and 0.35, respectively. The distribution of $PM_{2.1}/PM_{10}$ ratio is spatially similar to that of sulfate concentrations and the density of sulfur emissions.

Table 4 shows the day-to-day correlations between selected pollutants within each community. Correlations were calculated separately for summer days (May–September). When the warm season correlations differed from the annual correlation by more than 0.05, the values are shown in parentheses. The regional averages were calculated using the simple mean of individual community correlations. Annual and summer correlations between PM_{10} and $PM_{2.1}$, particle mass and sulfate con-

Table 3. Distribution of gaseous pollutant concentrations (ppb) by pollutant and community: 24 communities, United States and Canada, 1988–1991

Region	Samples[a]	Sulfur dioxide		Ammonia		Nitrous acid		Nitric acid	
		Mean	Max	Mean	Max	Mean	Max	Mean	Max
Sulfate Belt									
Hendersonville, TN	178	4.4	19.0	0.6	2.5	0.3	3.2	0.8	2.5
Oak Ridge, TN	172	3.9	20.7	0.4	3.6	0.5	3.3	0.8	2.0
Morehead, KY	173	4.5	56.0	0.6	4.2	0.6	3.3	0.9	3.5
Blacksburg, VA	172	5.5	18.8	1.2	12.3	0.6	2.5	0.8	4.3
Charlottesville, VA	167	3.4	22.2	0.3	1.4	0.1	1.8	0.9	15.7
Zanesville, OH	166	6.8	29.0	0.4	2.5	0.3	2.2	0.9	3.7
Athens, OH	180	10.9	39.1	0.5	1.8	0.6	2.5	1.0	3.4
Parsons, WV	162	6.2	32.8	0.5	2.7	0.4	3.5	0.7	2.4
Uniontown, PA	176	12.9	46.0	0.3	2.0	0.6	3.0	1.5	5.0
Penn Hill, PA	167	10.4	48.2	1.1	15.0	1.3	5.0	1.2	6.1
State College, PA	168	5.9	35.5	0.1	2.1	0.2	1.6	0.9	4.7
Transport Region									
Leamington, ON	175	4.9	25.1	1.0	10.6	0.4	1.5	0.9	4.0
Newtown, CT	161	4.0	23.4	0.7	20.8	0.6	4.5	0.6	4.4
Egbert, ON[b]	0								
Pembroke, ON	140	2.1	11.1	0.7	2.9	0.3	1.7	0.3	7.1
Dunnville, ON	139	5.2	18.7	1.3	5.7	0.3	2.8	1.0	4.9
South Brunswick, NJ[c]	48	5.9	20.0	3.5	11.9	1.0	2.7	1.4	3.5
West Coast									
Simi Valley, CA	169	0.7	2.5	3.2	10.8	1.2	5.2	2.1	12.2
Livermore, CA	179	1.0	8.3	2.7	16.1	1.4	7.4	0.9	12.7
Monterey, CA	176	0.3	1.0	0.7	4.4	0.3	2.6	0.4	2.0
Background									
Springdale, AR	164	1.1	4.3	5.8	28.5	0.5	2.3	0.3	1.1
Aberdeen, SD[d]	0								
Yorkton, SK[d]	0								
Penticton, BC	167	0.2	0.6	1.3	6.8	0.4	2.9	0.3	0.9

[a]Maximum sample size for any of the four parameters.
[b]Egbert sampling for 20 June 1990–2 December 1990.
[c]South Brunswick sampling for 21 June 1989–25 August 1989.
[d]Not measured at expected low-pollution sites.

centrations, and sulfate and hydrogen ion concentrations are all highly correlated in the Sulfate Belt and Transport Region. Summer correlations are higher among these parameters than for the year overall. The opposite appears true for the West Coast and Background sites.

Sulfate concentrations were higher in Sulfate Belt communities (Table 1): annual means ranged between 58 and 77 nmol/m^3 (6–8 μg/m^3). Summer means (Table 5) in the Sulfate Belt were 85–126 nmol/m^3 (9–13 μg/m^3), with the highest concentrations observed in Athens, Ohio; Uniontown and Penn Hills, Pennsylvania; and Morehead, Kentucky. The lowest concentrations were reported for the western sites, Penticton, British Columbia; Yorkton, Saskatchewan; and Monterey and Livermore, California. Maximum daily sulfate values were highest in western Pennsylvania and tended to be higher in the Sulfate Belt. Daily, summer, and annual sulfate concentrations decreased in all four directions away from the high sulfur emission area (*18*). In the Transport Region, summer sulfates were about two-thirds of the levels reported for the Sulfate Belt. Summer sulfate concentrations at the West Coast and Background sites were one-tenth to one-third of the concentrations in the Sulfate Belt.

Particulate nitrate concentrations (Fig. 2) were highest in Simi Valley (7.1 μg/m^3) and Livermore (5.3 μg/m^3). A few eastern sites had annual nitrate concentrations between 1 and 3.5 μg/m^3. The higher sites outside California were Leamington (3.4 μg/m^3) and Dunnville (2.3 μg/m^3), Ontario. The geographic patterns in the east for nitrates were consistent with those reported by Edgerton (*19*) from the EPA Dry Deposition Network in 1989. In that study, sites in the agricultural area of Illinois, Indiana, Ohio, and southern Michigan had nitrate concentrations in the range of 2–4 μg/m^3. Higher concentrations of nitrate occurred during the fall and winter period.

Particulate ammonium showed a more uniform pattern (Fig. 2) than nitrates over the eastern United States and Canada, with annual average concentrations between 1.5 and 2.5 μg/m^3. The higher concentrations were in western Pennsylvania and southern Ontario. Penticton, British Columbia, Monterey and Livermore, California, and Pembroke, Ontario had annual concentrations less than 1 μg/m^3. Summertime values in these four communities were equal to or less than the annual concentrations. In most cases, summer ammonium concentrations were 40–50% greater than the annual mean.

Ion balance ratios ($[H^+ + NH_4^+]/[2SO_4^{2-} + NO_3^-]$) indicate that particulate sulfate and nitrates are largely present in the form of ammonium salts. Adding these components together gives insight into the composition of fine particulate mass. Together ammonium sulfates and nitrates composed more than 50% of the $PM_{2.1}$ mass in 14 communities and more than 60% in 6 communities (Fig. 3). In those communities with the lowest percentage of sulfate and nitrate compounds composing $PM_{2.1}$ mass, in Morehead, Kentucky; Parson, West Virginia; and Pembroke, Ontario, more than half of the homes reported using wood as the primary heating fuel. Two other towns, Newtown, Connecticut, and Penticton, British Columbia, had wood burning in 34% and 40% of homes, respectively. Penticton, BC, and Monterey, California, both had low annual mean $PM_{2.1}$, about 9 μg/m^3 with 80% and 62%, respectively, composed of compounds other than sulfate and nitrate particles. The most likely contributors to fine particulate mass, other than ammonium sulfate and nitrate, are wood burning and motor vehicles. Analysis of elemental composition (bromine, potassium, etc.) and carbon (elemental and organic) is needed to resolve $PM_{2.1}$ source contribution.

Particle Strong Acidity

Particle strong acidity consists primarily of sulfuric acid (H_2SO_4) or partially neutralized ammonium bisulfate salts. Particle strong acidity is reported as hydrogen ion concentrations in nmol/m^3; the equivalent sulfuric acid concentration in μg/m^3 can be determined by dividing by 20.4.

The annual mean concentrations of particle strong acidity (Table 1) ranged from below the limit of detection in Yorkton, Saskatchewan, to approximately 50 nmol/m^3 in Uniontown, Pennsylvania, and Parsons, West Virginia. The summer or warm season (May–September) mean concentrations (Table 5) for most of the eastern United States and southern Canadian sites ranged between 50 and 90 nmol/m^3. These values were higher (1.6 to >2 times) than the annual mean concentrations. Particle strong acidity concentrations during the summer months exceeded 60 nmol/m^3 H^+ (3 μg/m^3 sulfuric acid equivalent) in a large geographic region that includes major por-

Table 4. Correlations of daily pollutant levels by pollutant and community, 24 communities, United States and Canada, 1988–1991

Region	$PM_{2.1}$ with PM_{10}	PM_{10} with sulfate particles	H^+ with sulfate particles	H^+ with ozone (1-hr maximum)	H^+ with nitric acid
Sulfate Belt					
Hendersonville, TN	0.58 (0.69)[a]	0.58 (0.65)	0.86	0.46 (0.26)	0.58 (0.66)
Oak Ridge, TN	0.85	0.63	0.89 (0.95)	0.32 (0.42)	0.41
Morehead, KY	0.80	0.78	0.88	0.55 (0.29)	0.50
Blacksburg, VA	0.82	0.78 (0.84)	0.89	0.64 (0.55)	0.44 (0.59)
Charlottesville, VA	0.87	0.79	0.93	0.46	0.09 (0.44)
Zanesville, OH	0.77	0.87	0.91	0.61	0.17
Athens, OH	0.90 (0.96)	0.79	0.93	0.59 (0.52)	0.63 (0.73)
Parsons, WV	0.79 (0.92)	0.54 (0.80)	0.95	0.57 (0.64)	0.40 (0.61)
Uniontown, PA	0.87	0.62	0.92	0.54	0.63 (0.72)
Penn Hill, PA	0.79	0.83	0.84	0.68 (0.62)	0.75
State College, PA	0.90 (0.95)	0.88 (0.94)	0.88	0.62	0.51 (0.64)
Combined	0.81	0.74	0.90	0.55	0.46
Transport Region					
Leamington, ON	0.78 (0.88)	0.74 (0.86)	0.83	0.52	0.56 (0.63)
Newtown, CT	0.78 (0.89)	0.72 (0.83)	0.88	0.68	0.70
Egbert, ON[b]	0.70 (0.77)	0.93	0.92	0.78	N/A
Pembroke, ON	0.74 (0.87)	0.70 (0.79)	0.89	0.53	0.35 (0.74)
Dunnville, ON	0.71	0.73	0.89	0.58	0.82
South Brunswick, NJ[c]	NA	NA	NA (0.71)	NA (0.52)	NA (0.49)
Combined	0.74	0.76	0.88	0.62	0.61
West Coast					
Simi Valley, CA	0.75	0.68 (0.49)	0.73 (0.45)	0.55 (0.42)	0.53
Livermore, CA	0.72 (0.64)	0.31 (0.21)	0.55	0.20 (0.33)	0.29 (0.37)
Monterey, CA	0.65 (0.33)	0.24 (0.00)	0.70	0.40	0.53 (0.40)
Combined	0.70	0.41	0.66	0.39	0.45
Background					
Springdale, AR	0.56 (0.48)	0.41	0.75	0.56 (0.35)	0.57 (0.46)
Aberdeen, SD[d]	0.41 (0.71)	0.01	0.62	0.45	NA
Yorkton, SK	0.33	0.14 (0.27)	0.26 (0.15)	0.07	NA
Penticton, BC	0.68	0.28	0.39	0.03 (0.16)	0.25 (0.09)
Combined	0.50	0.21	0.50	0.25	0.41

Abbreviations: $PM_{2.1}$, respirable particulate matter; PM_{10}, inhalable particulate matter; H^+, particle strong acidity; NA, not available.
[a]Summer correlations are presented in parentheses for those communities where summer (May–September) correlations differ from annual correlations by 0.05 or more.
[b]Egbert sampling for 21 June 1990–2 December 1990.
[c]South Brunswick sampling for 20 June 1989–25 August 1989.
[d]Aberdeen sampling for 29 May–September 1991.

Table 5. Summertime mean sulfate and mean particle strong acidity, 24 communities, United States and Canada, 1988–1991

Region	Sulfate (nmol/m³)	Particle strong acidity (nmol/m³)
Sulfate Belt		
Hendersonville, TN	100	64
Oak Ridge, TN	90	67
Morehead, KY	105	77
Blacksburg, VA	98	59
Charlottesville, VA	85	64
Zanesville, OH	103	65
Athens, OH	104	77
Parsons, WV	90	82
Uniontown, PA	108	88
Penn Hills, PA	126	69
State College, PA	94	67
Transport Region		
Leamington, ON	66	30
Newtown, CT	66	42
Egbert, ON	49	11
Pembroke, ON	46	29
Dunnville, ON	94	54
South Bruswick, NJ	72	48
West Coast		
Simi Valley, CA	42	19
Livermore, CA	17	11
Monterey, CA	14	9
Background		
Springdale, AR	50	21
Aberdeen, SD	25	4
Yorkton, SK	8	0
Penticton, BC	7	9

tions of Tennessee, Kentucky, Ohio, Virginia, Pennsylvania, New York, and West Virginia. Single day concentrations throughout the Sulfate Belt area exceeded 150 nmol/m³ (7.5 μg/m³) (Fig. 3). In seven communities there were days when particle strong acidity concentrations exceeded 500 nmol/m³ (25 μg/m³). Transport events were responsible for highly variable exposures in the northeast, with occasional high particle strong acidity exposure days, but a fairly low annual mean.

The correlation between sulfate and PM_{10} daily concentrations was higher in the Sulfate Belt and Transport Region than in the other communities (Table 4). Among the eastern communities in the high sulfate region, the daily sulfate to particle strong acidity correlations averaged 0.9 annually and for the summer months. Strong correlations were also observed for communities affected by transported acid sulfates (r = 0.88). These findings suggest that daily variations in particle mass, sulfate, and acidity measurements are all highly correlated in the Sulfate Belt and Transport Regions of North America.

Gaseous Pollutants

Annual mean ozone concentrations ranged from 16 ppb in Yorkton, Saskatchewan, to 35 ppb in Simi Valley, California; there was a slightly greater than twofold difference across the range of communities (Table 2). The interquartile range of annual mean concentrations was 7 ppb. There was a threefold range of 8-hr daytime ozone concentrations (Table 2).

The United States and Canadian health standards for ozone are based on acute 1-hr exposures. The Canadian 1-hr ozone standard, 100 ppb, was exceeded in 18 communities; the United States standard, 0.12 ppm, was exceeded in 10 communities (Table 2). Simi Valley, just north of Los Angeles, had the second highest mean for the daily maximum hourly and the second highest single hourly value recorded, 170 ppb, behind Newtown, Connecticut. The same was true for the daytime (1000–1800 hr) averaged concentrations but not for the 24-hr mean concentrations. Annual average ozone concentrations in Hendersonville, Tennessee (20 km northeast of Nashville) and the more rural locations of Morehead, Kentucky; Dunnville, Ontario; and State College, Pennsylvania, approached those in Simi Valley. In fact, the maximum 24-hr average ozone concentration in five of the eastern communities exceeded the maximum 77 ppb 24-hr mean observed in Simi Valley. Ozone concentrations in Simi Valley displayed the greatest diurnal variation, particularly during the summer months.

The correlation between daily particle strong acidity concentrations and maximum 1-hr ozone levels did not show any distinct patterns (Table 4). The summer and annual correlations were similar for the most part. The only exceptions were in Hendersonville, Tennessee; and Morehead, Kentucky, where the summer correlations are substantially lower than other sites in

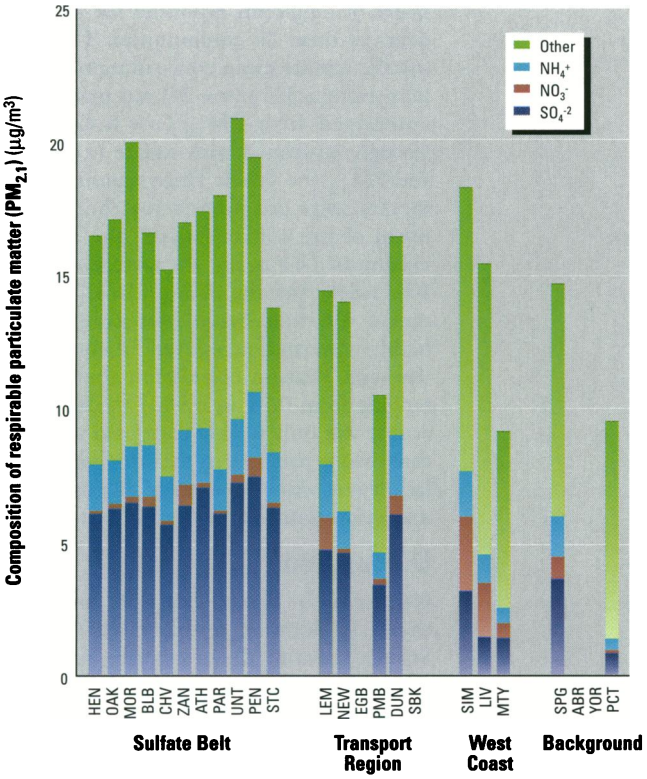

Figure 2. Composition of respirable matter (µg/m³): 24 cities, United States and Canada, 1988–1991. See Table 1 for site codes.

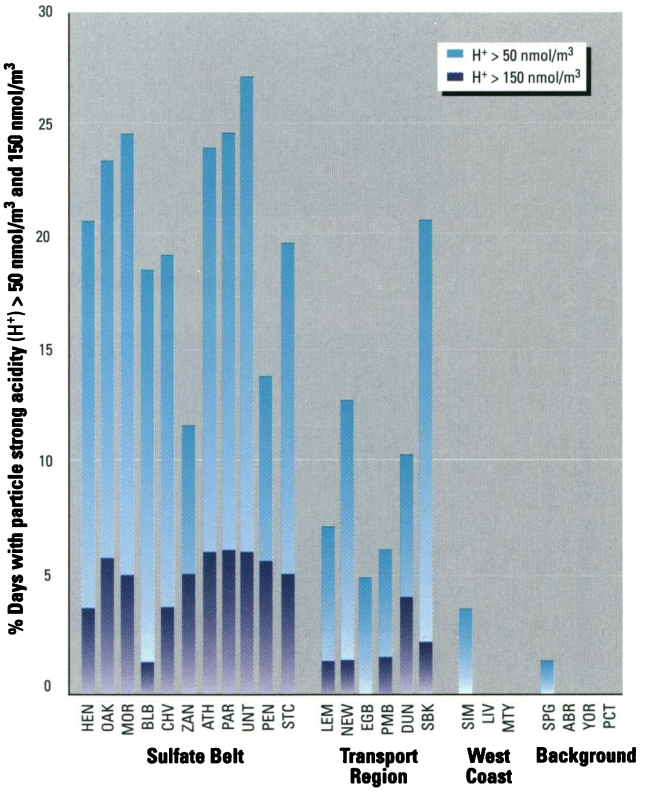

Figure 3. Percent of days with particle strong acidity concentrations >50 nmol/m³ and 150 nmol/m³: 24 cities, United States and Canada, 1988–1991. See Table 1 for site codes.

the east. The daily correlation of ozone with particle strong acidity was low in Hendersonville (r = 0.26) and in Morehead (r = 0.29), while for the other communities in the Sulfate Belt daily correlations of ozone with particle strong acidity were higher than 0.60. Parkhurst et al. (*20*) have reported that ozone in Hendersonville is substantially and consistently higher than levels measured in Nashville and higher than other regional Tennessee Valley Authority monitoring sites. These observations suggest that the southwest winds transporting ozone precursors from Nashville lead to elevated ozone concentrations. However, high concentrations of particle strong acidity are not associated with these meteorological conditions.

Ammonia is associated with natural, industrial, and agricultural sources. The highest mean ammonia concentrations (Table 3) were observed in Springdale, Arkansas, a community with a high density of chicken farms and processing plants. The farming communities of Leamington and Dunnville, Ontario; Blacksburg, Virginia; and Penticton, British Columbia, also had annual mean ammonia concentrations exceeding 1 ppb, as did Penn Hills, Pennsylvania, an eastern suburb of the industrial city of Pittsburgh (population 2.2 million).

For the eastern sites, nitric acid ranged between 0.7 and 1.5 ppb (29–61 nmol/m³; Table 3). The nitric acid annual mean concentration in Simi Valley, California, was more than double the mean concentration for almost all other sites. Ten percent of days in Simi Valley had concentrations exceeding 4.3 ppb, with the highest single day of 12.2 ppb. Annual nitric acid concentrations exceeded 1 ppb only in Uniontown and Penn Hills, Pennsylvania. At most sites the summer concentrations were equal to or greater than the annual mean. However, Edgerton et al. (*18*) observed that winter nitric acid concentrations were higher than other seasons during 1989 across the National Dry Deposition Network.

The highest annual nitrous acid concentration (Table 3) was in Livermore, California (1.4 ppb or 57 nmol/m³), followed by Penn Hills, Pennsylvania (1.3 ppb or 53 nmol/m³) and Simi Valley, California (1.2 ppb or 49 nmol/m³). Nitrous acid concentrations were about one-third the nitric acid concentrations except at these three sites. There was more gas-phase acidity (nitric and nitrous acid) than particle-phase strong acidity at every site except Parsons, West Virginia. There was eight times more gas-phase acidity than particle-phase acidity in Livermore and Simi Valley. But, even downwind of Pittsburgh, the Penn Hills site had substantially more gas-phase acidity than particle-phase acid (102 nmol/m³ versus 30 nmol/m³).

Seasonal Patterns in Particle Strong Acidity

Monthly mean concentrations of particle strong acidity in the eastern communities were elevated during May–September, whereas in the western sites of Simi Valley and Monterey, California, and Penticton, British Columbia, there was little seasonal variability (Fig. 4). Particle strong acidity occurs during meteorological conditions that favor the conversion of sulfur dioxide to acid sulfates without the presence of excess ammonia. Although these meteorological conditions occur more frequently during summer months, they can occur in other seasons. In the more southern towns of Hendersonville, Tennessee, and Morehead, Kentucky, elevated particle strong acidity was observed during the winter months. Particle strong acidity in Uniontown, Pennsylvania, was elevated during December, April, and May, as well as during the summer months (Fig. 4). The elevated monthly mean for December was the result of a single event when the 24-hr concentration reached 133 nmol/m³, possibly due to the impact of a nearby coal-fired power plant.

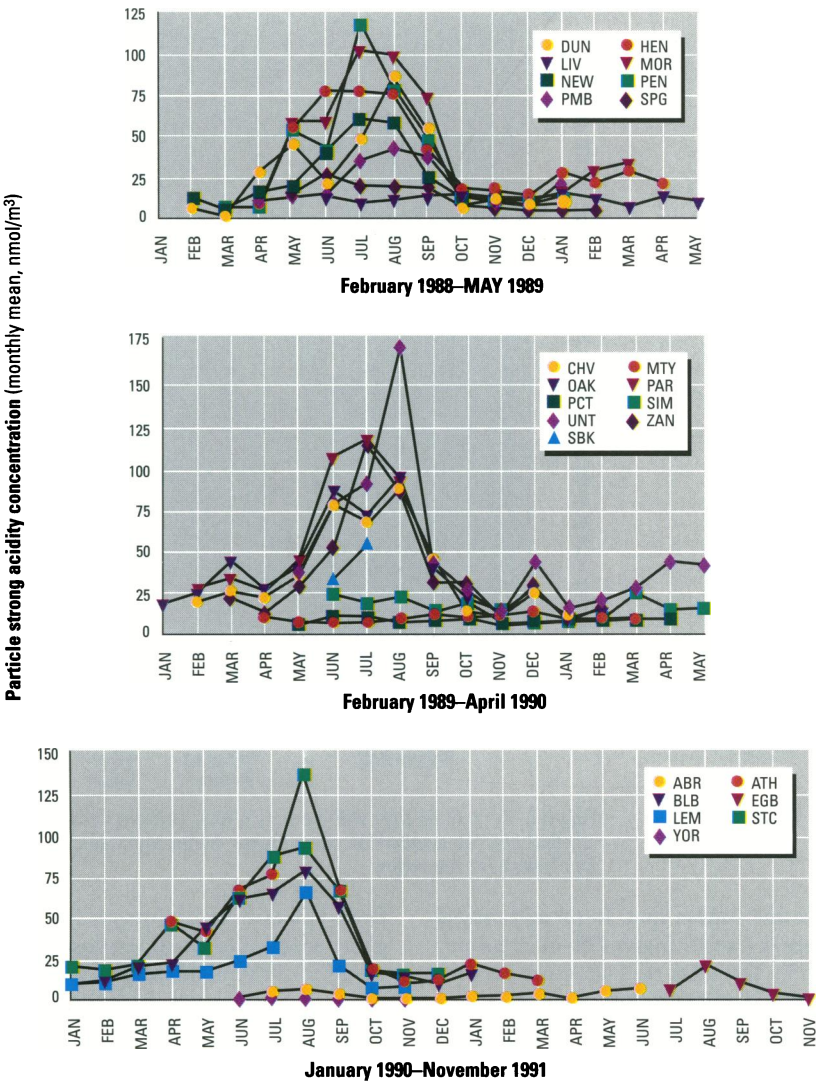

Figure 4. Monthly mean concentrations of particle strong acidity (nmol/m³): 24 cities, United States and Canada, 1988–1991. See Table 1 for site codes.

The cumulative monthly percent contribution to the total annual exposure was calculated for each community. With the exception of Livermore, California, 10–20% of the acid occurred during the first 4 months of the year. Less than 20% occurred during the last three months. In the Sulfate Belt and Transport Regions, between 60 and 80% of the annual particle strong acidity exposure occurs between the beginning of May and the end of September. In contrast, in the western communities the particle strong acidity exposures are evenly distributed throughout the year.

The particle strong acidity events data from Oak Ridge, Tennessee; Parsons, West Virginia; Penn Hills, Pennsylvania; and Pembroke, Ontario—the four communities on an axis through the highest particle strong acidity concentrations—were closely examined (*21*). Days were ranked by particle acidity, and the cumulative annual exposure was normalized to 100%. For comparison, the number of days required to reach 75% of the annual particle strong acidity exposure is an indication of the episodic nature of particle strong acidity events. The community farthest south, Oak Ridge, reached 75% of the annual exposure in 40% of the days. The communities to the northeast received 75% of the annual particle strong acidity exposures in less than 4 months (33%) of the year. In Penn Hills, proximate to Pittsburgh, only 22% of the days were required to reach 75% of the annual exposure. Although particle strong acidity events can occur throughout the year, particle strong acidity pollution is episodic and more likely during spring and summer months.

Interrelationships of Annual Means

The annual mean concentrations of these pollutants (Tables 1 and 2) provide estimates of long-term exposures for the residents of these 24 communities. The city-specific annual mean concentration of particle strong acidity (n = 24) was moderately correlated with PM_{10} (r = 0.47), but strongly correlated with sulfate (r = 0.90) and $PM_{2.1}$ (r = 0.82). Three exposure parameters were considered for the annual mean ozone concentration: the average maximum 1-hr mean, the average daytime 8-hr mean, and the average daily 24-hour mean. All three ozone parameters were highly correlated across the 22 communities with Pearson correlation coefficients ranging from 0.74 to 0.98. Particle strong acidity was only weakly correlated with the three ozone parameters; the strongest correlation was with the average daytime 1-hr maximum ozone concentration (r = 0.37).

Discussion

Neither sulfur dioxide nor PM_{10} exceeded the U.S. National Ambient Air Quality Standards during the monitoring period in any of the study communities. Sulfur dioxide concentrations reached 56 ppb on the highest single day, which is 40% of the 24-hr standard. The highest annual mean was 13 ppb, about one-third of the standard. Inhalable particulate matter (PM_{10}) annual concentrations were all less than two-thirds of the United States standards of 50 μg/m³ annual mean. There were no recorded exceedences of the 150 μg/m³ maximum daily standard. Across the Sulfate Belt and Transport Regions, fine particles ($PM_{2.1}$) were 60% to more than 80% of the mean PM_{10} mass concentration. Both particle mass measures ($PM_{2.1}$ and PM_{10}) and the sulfate concentrations were highly correlated. Similarly, high correlations were observed in these two regions during the warm season between particle strong acidity and the particle measurements ($PM_{2.1}$, PM_{10}, and sulfate).

Annual and daytime mean ozone concentrations varied by only a factor of two or three, respectively, across sites. Although the site selection did provide a contrast among clean communities (low particles, acidity, and ozone) and communities with higher air pollution, it proved difficult to find sites with high particle strong acidity but low ozone. The West Coast communities of Simi Valley and Livermore, California, had higher ozone concentrations with low particle sulfate and hence low particle strong acidity. However, these two communities had substantial gas-phase acidity.

There was a 10-fold difference in atmospheric sulfate and a larger range of acid particle concentrations. The higher annual, seasonal (spring and summer), and daily particle strong acidity concentrations

occurred in communities along the western edge of the Appalachian Plateau and Allegheny Mountains. Days occurred when the atmospheric particle strong acidity exceeded 300 nmol/m^3 (15 μg/m^3 sulfuric acid equivalent) throughout the Sulfate Belt and into the downwind Transport Region in Canada and the eastern United States. Annual mean community exposures are determined, in part, by the number of particle strong acidity events. Days with elevated particle strong acidity occurred over highly populated urban and rural areas of the eastern United States.

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